

Amendments to the Claims

Please add Claim 59. The Claim Listing below will replace all prior versions of the claims in the application:

Claim Listing

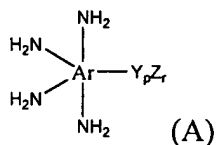
1-28. (Cancelled)

29. (Previously Presented) A proton-conducting polymer membrane which comprises polyazoles containing phosphonic acid groups and is obtainable by a process comprising the steps:

- A) mixing one or more aromatic or heteroaromatic tetraamino compounds with one or more aromatic or heteroaromatic carboxylic compounds in polyphosphoric acid to form a solution or dispersion, wherein the carboxylic compounds contain at least two carboxylic groups selected from acids, esters, acid halides or acid anhydrides, with at least part of the tetraamino compounds or the carboxylic compounds comprising at least one phosphonic acid group; or
mixing one or more aromatic or heteroaromatic diaminocarboxylic compounds in polyphosphoric acid to form a solution or dispersion, wherein the diaminocarboxylic compounds contain a carboxylic group selected from acids and esters, wherein at least a part of said diaminocarboxylic compounds comprises phosphonic acid groups;
- B) heating the solution or dispersion obtained according to step A) under inert gas to temperatures of up to 350°C to form polyazole polymers; and
- C) applying a layer using the mixture from step B) to a support, thus forming a membrane on the support; or
- D) applying the solution or dispersion from step A) to a support, thus forming a membrane on the support; and
- E) heating the membrane formed in step D) under inert gas to temperatures of up to 325°C to form polyazole polymers; and

F) partially hydrolyzing the polyphosphoric acid moieties of the membrane from step C) or step E) until the membrane is self-supporting.

30. (Previously presented) The membrane of Claim 29, characterized in that the mixture prepared in step A) comprises aromatic or heteroaromatic tetraamino compounds of the formula (A)



where

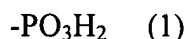
Ar is an aromatic or heteroaromatic group,

Y is a bond or a group having from 1 to 20 carbon atoms,

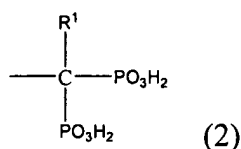
p is an integer from 1 to 4 and represents the number of bonds or groups Y via which the group Z is bound to the group Ar,

r is an integer from 1 to 4 and represents the number of groups Z which are bound to the group Y or, if Y is a bond, to the aromatic or heteroaromatic group Ar, and

Z is a group of the general formula (1)

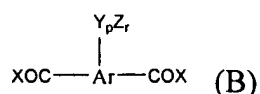


or the general formula (2)



where R^1 is a hydrogen atom or a group having from 1 to 20 carbon atoms.

31. (Previously Presented) The membrane of Claim 29, characterized in that the mixture prepared in step A) comprises aromatic or heteroaromatic carboxylic compounds of the formula (B)



where

Ar is an aromatic or heteroaromatic group,

X is a halogen atom or a group of the formula OR^2 , where R^2 is a hydrogen atom or a group having from 1 to 20 carbon atoms,

Y is a bond or a group having from 1 to 20 carbon atoms,

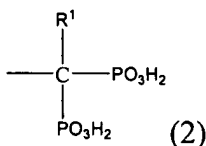
p is an integer from 1 to 4 and represents the number of bonds or groups Y via which the group Z is bound to the group Ar,

r is an integer from 1 to 4 and represents the number of groups Z which are bound to the group Y or, if Y is a bond, to the aromatic or heteroaromatic group Ar, and

Z is a group of the general formula (1)

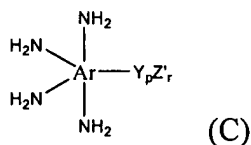


or the general formula (2)



where R^1 is a hydrogen atom or a group having from 1 to 20 carbon atoms.

32. (Previously presented) The membrane of Claim 29, characterized in that the mixture prepared in step A) comprises aromatic or heteroaromatic tetraamino compounds of the formula (C)



where

Ar is an aromatic or heteroaromatic group,

Y is a bond or a group having from 1 to 20 carbon atoms,

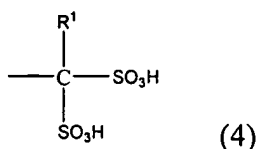
p is an integer from 1 to 4 and represents the number of bonds or groups Y via which the group Z is bound to the group Ar,

r is an integer from 1 to 4 and represents the number of groups Z which are bound to the group Y or, if Y is a bond, to the aromatic or heteroaromatic group Ar, and

Z' is a group of the general formula (3)

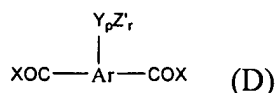


or the general formula (4)



where R¹ is a hydrogen atom or a group having from 1 to 20 carbon atoms.

33. (Previously Presented) The membrane of Claim 29, characterized in that the mixture prepared in step A) comprises aromatic or heteroaromatic carboxylic compounds of the formula (D)



where

Ar is an aromatic or heteroaromatic group which optionally bears further substituents,

X is a halogen atom or a group of the formula OR², where R² is a hydrogen atom or a group having from 1 to 20 carbon atoms,

Y is a bond or a group having from 1 to 20 carbon atoms,

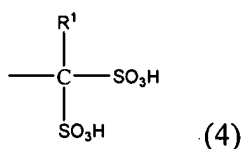
p is an integer from 1 to 4 and represents the number of bonds or groups Y via which the group Z is bound to the group Ar,

r is an integer from 1 to 4 and represents the number of groups Z which are bound to the group Y or, if Y is a bond, to the aromatic or heteroaromatic group Ar, and

Z' is a group of the general formula (3)



or the general formula (4)



where R¹ is a hydrogen atom or a group having from 1 to 20 carbon atoms.

34. (Previously Presented) The membrane of Claim 29, characterized in that the mixture prepared in step A) comprises aromatic or heteroaromatic tetraamino compounds which contain no phosphonic acid groups and aromatic or heteroaromatic carboxylic acids compounds which contain at least one phosphonic acid group.
35. (Previously presented) The membrane of Claim 29, characterized in that the mixture prepared in step A) comprises 3,3',4,4'-tetraaminobiphenyl, 2,3,5,6-tetraaminopyridine, or 1,2,4,5-tetraaminobenzene.
36. (Previously Presented) The membrane of Claim 29, characterized in that the mixture prepared in step A) comprises isophthalic acid, terephthalic acid, phthalic acid, 5-hydroxyisophthalic acid, 4-hydroxyisophthalic acid, 2-hydroxyterephthalic acid, 5-aminoisophthalic acid, 5-N,N-dimethylaminoisophthalic acid, 5-N,N-diethylaminoisophthalic acid, 2,5-dihydroxyterephthalic acid, 2,5-dihydroxyisophthalic acid, 2,3-dihydroxyisophthalic acid, 2,3-dihydroxyphthalic acid, 2,4-dihydroxyphthalic acid, 3,4-dihydroxyphthalic acid, 3-fluorophthalic acid, 5-fluoroisophthalic acid, 2-fluoroterephthalic acid, tetrafluorophthalic acid, tetrafluoroisophthalic acid, tetrafluoroterephthalic acid, 1,4-naphthalenedicarboxylic acid, 1,5-naphthalenedicarboxylic acid, 2,6-naphthalenedicarboxylic acid, 2,7-naphthalenedicarboxylic acid, diphenic acid, 1,8-dihydroxynaphthalene-3,6-dicarboxylic acid, bis(4-carboxyphenyl) ether, benzophenone-4,4'-dicarboxylic acid, bis(4-dicarboxyphenyl) sulfone, biphenyl-4,4'-dicarboxylic acid, 4-trifluoromethylphthalic acid, 2,2-bis(4-carboxyphenyl)hexafluoropropane, 4,4'-stilbenedicarboxylic acid, 4-carboxycinnamic acid, or their C1-C20-alkyl esters or C5-C12-aryl esters, or their acid anhydrides or acid chlorides.
37. (Previously Presented) The membrane of Claim 29, characterized in that the mixture prepared in step A) comprises 2,3-diamino-5-carboxyphenylphosphonic acid, 2,3-

diamino-6-carboxyphenylphosphonic acid, and 3,4-diamino-6-carboxyphenylphosphonic acid.

38. (Previously presented) The membrane of Claim 29, characterized in that the mixture prepared in step A) comprises 2,3-diamino-5-carboxyphenylsulfonic acid, 2,3-diamino-6-carboxyphenylsulfonic acid, and 3,4-diamino-6-carboxyphenylsulfonic acid.
39. (Previously Presented) The membrane of Claim 29, characterized in that the mixture prepared in step A) comprises aromatic tricarboxylic compounds selected from tricarboxylic acids, their C1-C20-alkyl esters or C5-C12-aryl esters or their acid anhydrides or their acid halides or tetracarboxylic acids, their C1-C20-alkyl esters or C5-C12-aryl esters or their acid anhydrides or their acid halides.
40. (Previously presented) The membrane of Claim 39, characterized in that the mixture prepared in step A) comprises: 1,3,5-benzenetricarboxylic acid (trimesic acid), 2,4,5-benzenetricarboxylic acid (trimellitic acid), (2-carboxyphenyl)iminodiacetic acid, 3,5,3'-biphehyltricarboxylic acid, 3,5,4'-biphenyltricarboxylic acid, 2,4,6-pyridinetricarboxylic acid, benzene-1,2,4,5-tetracarboxylic acid, naphthalene-1,4,5,8-tetracarboxylic acid, 3,5,3',5'-biphenyltetracarboxylic acid, benzophenonetetracarboxylic acid, 3,3',4,4'-biphenyltetracarboxylic acid, 2,2',3,3'-biphenyltetracarboxylic acid, or 1,2,5,6-naphthalenetetracarboxylic acid.
41. (Previously Presented) The membrane of Claim 39, characterized in that the content of tricarboxylic compound or tetracarboxylic compound is in the range of from 0 to 30 mol % based on dicarboxylic acid used.
42. (Previously Presented) The membrane of Claim 41, characterized in that the content of tricarboxylic compound or tetracarboxylic compound is in the range of from 0.1 to 20 mol % based on dicarboxylic acid used.

43. (Previously Presented) The membrane of Claim 42, characterized in that the content of tricarboxylic compound or tetracarboxylic compound is in the range of from 0.5 to 10 mol % based on dicarboxylic acid used.
44. (Previously Presented) The membrane of Claim 29, characterized in that the mixture prepared in step A) comprises heteroaromatic dicarboxylic compound, tricarboxylic compound or tetracarboxylic compound which contain at least one nitrogen, oxygen, sulfur, or phosphorus atom in the aromatics.
45. (Previously presented) The membrane of Claim 44, characterized in that pyridine-2,5-dicarboxylic acid, pyridine-3,5-dicarboxylic acid, pyridine-2,6-dicarboxylic acid, pyridine-2,4-dicarboxylic acid, 4-phenyl-2,5-pyridinedicarboxylic acid, 3,5-pyrazoledicarboxylic acid, 2,6-pyrimidinedicarboxylic acid, 2,5-pyrazinedicarboxylic acid, 2,4,6-pyridinetricarboxylic acid, benzimidazole-5,6-dicarboxylic acid or their C1-C20-alkyl esters or C5-C12-aryl esters or their acid anhydrides or their acid chlorides are used.
46. (Previously presented) The membrane as claimed in claim 29, characterized in that the mixture prepared in step A) comprises diaminobenzoic acid or its monohydrochloride and dihydrochloride salts.
47. (Cancelled)
48. (Previously Presented) The membrane as claimed in claim 29, characterized in that the solution produced in step A) further comprises dispersed or suspended polymer.
49. (Previously Presented) The membrane of Claim 29, characterized in that the treatment according to step F) is carried out at temperatures in the range of from 0°C to 150°C in the presence of moisture.

50. (Previously Presented) The membrane of Claim 29, characterized in that the treatment of the membrane in step F) is carried out from 10 seconds to 300 hours.
51. (Previously Presented) The membrane of Claim 29, characterized in that the membrane formed after step F) is crosslinked by action of oxygen.
52. (Previously Presented) The membrane of Claim 29, characterized in that a layer having a thickness of from 20 to 4000 μm is produced in step C) or step D).
53. (Previously Presented) The membrane of Claim 29, characterized in that the membrane formed after step F) has a thickness of from 15 to 3000 μm .
54. (Previously Presented) An electrode having a proton-conducting polymer coating which is based on polyazoles and is obtainable by a process comprising the steps:
- A) mixing one or more aromatic or heteroaromatic tetraamino compounds with one or more aromatic or heteroaromatic carboxylic acids compounds in polyphosphoric acid to form a solution or dispersion, wherein the carboxylic compounds contain at least two groups selected from acids, esters, acid halides or acid anhydrides, with at least part of the tetraamino compounds or the carboxylic compounds comprising at least one phosphonic acid group; or
mixing one or more aromatic or heteroaromatic diaminocarboxylic compounds in polyphosphoric acid to form a solution or dispersion, wherein the diaminocarboxylic compounds contain a group selected from acids and esters, wherein at least a part of said diaminocarboxylic compounds comprises phosphonic acid groups;
 - B) heating the solution or dispersion obtained according to step A) under inert gas to temperatures of up to 350°C to form the polyazole polymer; and
 - C) applying a layer using the mixture from step B) to an electrode, thus forming a membrane on the electrode; or

- D) applying the solution or dispersion from step A) to an electrode, thus forming a membrane on the electrode; and
 - E) heating the membrane formed in step D) under inert gas to temperatures of up to 325°C to form polyazole polymers; and
 - F) partially hydrolyzing the polyphosphoric acid moieties of the electrode membrane formed in step C) or step E) until the membrane has a surface hardness.
55. (Previously Presented) The electrode of Claim 54, wherein the membrane has a thickness of from 2 to 3000 μm .
56. (Previously presented) A membrane-electrode unit comprising at least one electrode and at least one membrane as claimed in Claim 29.
57. (Previously Presented) A membrane-electrode unit comprising at least one electrode having a proton-conducting polymer coating which is based on polyazoles and is prepared by the following steps:
- A) mixing one or more aromatic or heteroaromatic tetraamino compounds with one or more aromatic or heteroaromatic carboxylic compounds in polyphosphoric acid to form a solution or dispersion, wherein the carboxylic compounds contain at least two groups selected from acids, esters, acid halides or acid anhydrides, with at least part of the tetraamino compounds or the carboxylic compounds comprising at least one phosphonic acid group; or
mixing one or more aromatic or heteroaromatic diaminocarboxylic compounds in polyphosphoric acid to form a solution or dispersion, wherein the diaminocarboxylic compounds contain a group selected from acids and esters, wherein at least a part of said diaminocarboxylic compounds comprises phosphonic acid groups;
 - B) heating the solution or dispersion obtained according to step A) under inert gas to temperatures of up to 350°C to form the polyazole polymer; and

- C) applying a layer using the mixture from step B) to an electrode, thus forming a membrane on the electrode; or
 - D) applying a solution or dispersion from step A) to an electrode, thus forming a membrane on the electrode; and
 - E) heating the membrane formed in step D) under inert gas to temperatures of up to 325°C to form polyazole polymers; and
 - F) partially hydrolyzing the polyphosphoric acid moieties of the electrode membrane formed in step C) until the membrane has a surface hardness.
58. (Previously presented) A fuel cell comprising one or more membrane-electrode units as claimed in Claim 57.
59. (New) A proton-conducting polymer membrane which comprises polyazoles containing phosphonic acid groups and is obtainable by a process comprising the steps:
- A) mixing one or more aromatic or heteroaromatic tetraamino compounds with one or more aromatic or heteroaromatic carboxylic compounds in polyphosphoric acid to form a solution or dispersion, wherein the carboxylic compounds contain at least two carboxylic groups selected from acids, esters, acid halides or acid anhydrides, with at least part of the tetraamino compounds or the carboxylic compounds comprising at least one phosphonic acid group; or
mixing one or more aromatic or heteroaromatic diaminocarboxylic compounds in polyphosphoric acid to form a solution or dispersion, wherein the diaminocarboxylic compounds contain a carboxylic group selected from acids and esters, wherein at least a part of said diaminocarboxylic compounds comprises phosphonic acid groups;
 - B) heating the solution or dispersion obtained according to step A) under inert gas to temperatures of up to 350°C to form polyazole polymers; and
 - C) applying a layer using the mixture from step B) to a support, thus forming a membrane on the support; or

- D) applying the solution or dispersion from step A) to a support, thus forming a membrane on the support; and
- E) heating the membrane formed in step D) under inert gas to temperatures of up to 325°C to form polyazole polymers; and
- F) partially hydrolyzing the polyphosphoric acid moieties of the membrane from step C) or step E) until the membrane is self-supporting,

wherein the concentration of phosphoric acid in the membrane of step (F) is from 10 to 80 mols of phosphoric acid per mol of a repeating unit of the polyazole polymer.